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Results of time-of-flight transmission measurements for ^{nat}Ce samples at GELINA

Description of GELINA data to be stored in the EXFOR data base

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Results of time-of-flight transmission measurements for ^{nat}Ce samples at GELINA

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Contents

Abstract

Measurements have been performed at the time-of-flight facility GELINA to determine the total cross section for neutron induced reactions in ^{nat}Ce. The measurements have been carried out at the 50 m transmission station at a moderated neutron beam using a Li-glass scintillator with the accelerator operating at 800 Hz. This report describes the experimental details required to deliver the data to the EXFOR data library which is maintained by the Nuclear Data Section of the IAEA and the Nuclear Energy Agency of the OECD. The experimental conditions and data reduction procedures are described and results are compared to evaluated data. In addition, the full covariance information based on the AGS concept is given such that nuclear reaction model parameters together with their covariances can be derived in a least squares adjustment to the data.

1 Introduction

To study the resonance structure of neutron induced reaction cross sections, neutron spectroscopic measurements are required which determine with a high accuracy the energy of the neutron that interacts with the material under investigation. To cover a broad energy range such measurements are best carried out with a pulsed white neutron source, which is optimized for time-of-flight (TOF) measurements [1].

The TOF facility GELINA [2] has been designed and built for high-resolution cross section measurements in the resolved (RRR) and unresolved (URR) resonance region. It is a multi-user TOF facility, providing a white neutron source with a neutron energy range from 10 meV to 20 MeV. Up to 10 experiments can be performed simultaneously at measurement stations located between 10 m to 400 m from the neutron production target. The electron linear accelerator provides a pulsed electron beam with a maximum energy of 150 MeV, a peak current of 10 A and a repetition rate ranging from 50 Hz to 800 Hz. A compression magnet reduces the width of the electron pulses to less than 1 ns [3]. The electron beam hits a mercury-cooled uranium target producing Bremsstrahlung and subsequently neutrons via photonuclear reactions [4]. Two water-filled beryllium containers mounted above and below the neutron production target are used to moderate the neutrons. By applying different neutron beam collimation conditions, experiments can use either a fast or a moderated neutron spectrum. The neutron production rate is constantly monitored by BF_3 proportional counters which are mounted in the ceiling of the target hall. The output of the monitors is used to normalize the time-of-flight spectra to the same neutron intensity. The measurement stations are equipped with air conditioning to reduce electronic drifts in the detection chains due to temperature changes.

In this report results of transmission measurements carried out at GELINA with two natural cerium metal samples provided by the Oak Ridge National Laboratory are described. To reduce bias effects due to e.g. dead time and background, the measurement and data reduction procedures recommended in Ref. [1] have been followed. The main objective of this report is to provide the information that is required to evaluate the total cross section for cerium isotopes in the resonance region and to extract nuclear reaction model parameters in a least squares adjustment to the data. The recommendations resulting from a consultant's meeting organized by the Nuclear Data Section of the IAEA are followed in the description of the data [5].

2 Experimental conditions

The measurements reported in this work were performed at 50 m transmission measurement station of flight path 4 of GELINA. This flight path forms an angle of 9° with respect to the normal of the moderator surface facing the flight path. The accelerator was operated at 800 Hz and produced an average beam current of about 55 µA. A shadow bar made of copper and lead was placed close to the uranium target to reduce the intensity of both the γ-ray flash and the fast neutron component. The samples and the detector were placed in climatised rooms at a constant temperature of 22°C. A schematic view of the experimental set-up is shown in Figure 1. The experimental conditions, including the facility, the sample, the detector characteristics and the type of background and overlap filters used in the experiment, are specified in Appendix A.

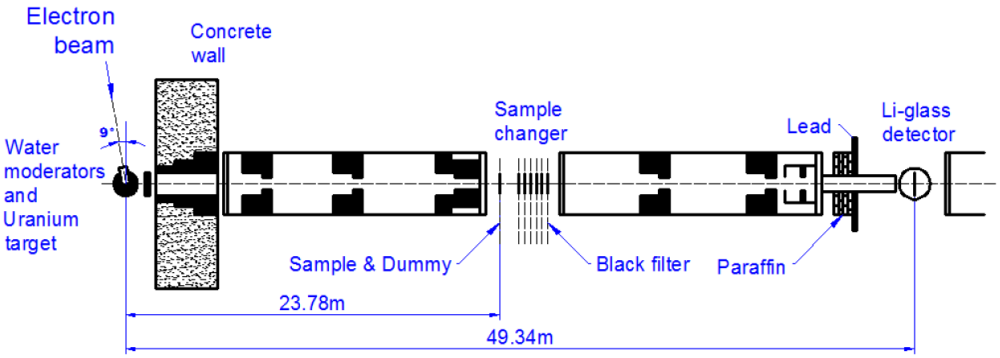

Figure 1 Schematic representation of the transmission set-up at the 50 m transmission station of GELINA.

A collimation system, mainly composed of lithium-carbonate plus resin, copper and lead collimators, reduced the neutron beam to a diameter of about 45 mm at the sample position. The sample was placed in an automatic sample changer at a distance of approximately 24 m from the neutron source. To minimize the contribution of neutrons from previous bursts a ^{10}B overlap filter with an areal density of about 0.08 at/b was placed in the beam close to the neutron target. The impact of the γ-ray flash was reduced by a 16 mm thick Pb filter. Additional black resonance filters (Na, Co and W) were mounted in the automatic and independent filter changers close to the sample position to determine the background with the black resonance technique [1].

The neutron passing through the sample and filters was further collimated and detected by a 6.35 mm thick and 151.6 mm diameter NE912 Li-glass scintillator. The scintillator was connected through a boron-free quartz window to a 127 mm EMI 9823 KQB photomultiplier (PMT), which was placed outside the neutron beam perpendicular to its axis. The diameter of the neutron beam at the detector position was about 90 mm.

The output signals of the detector were connected to conventional analog electronics. The anode pulse of the PMT was fed into a constant fraction discriminator to create a fast logic signal which defines the time the neutron has been detected. The signal of the 9th dynode was shaped by a spectroscopic amplifier to determine the energy deposited by the ^{6}Li(n,t)α reaction in the detector. A module was included to produce a fixed dead time in the whole electronics chain directly after the detection of an event. This dead time t_d = 3305 (10) ns was continuously monitored by recording the time interval between successive pulses. The time-of-flight (TOF) of the detected neutron was determined by the time difference between the start signal (T_0), given at each electron burst, and the stop signal (T_s) derived from the anode pulse of the PMT. This time

difference was measured with a multi-hit fast time coder with a 1 ns time resolution [7]. The TOF and pulse height of a detected event were recorded in list mode using a multi-parameter data acquisition system developed at the JRC-Geel [7]. Each measurement was subdivided in different cycles alternating different configurations with and without sample and filters in beam. Only cycles for which the ratio between the total counts in the transmission detector and in the neutron beam monitor deviated by less than 1% from the average value were selected. This selection was done with the code described in Ref. [8].

Measurements were carried out with natural metallic cerium samples which were encapsulated in a thin-walled aluminium can to prevent reaction with air. Two disks with a diameter of 59.85 mm and a respective thickness of 1.85 mm and 9.93 mm (see Fig. 1) were produced at the Oak Ridge National Laboratory. The weight of the disks was 34.975 g and 187.945 g, respectively. To compensate for the effect of the metallic can, additional empty containers for both samples were manufactured and measured in the same beam conditions. Detailed sample characteristics are given in Table 1.

Sample (mm)	Shape	Area/cm^2	Mass/g	Areal Density (at/b)	Station (filters)
^{nat}Ce (1.85)	Disk	28.14 (0.01)	34.975 (0.015)	5.534 (0.025) x10^{-3}	50 m (Co, W)
^{nat}Ce (9.93)	Disk	28.13 (0.01)	187.954 (0.015)	28.713 (0.025) x10^{-3}	50 m (Co, W)

Table 1 Characteristics of the samples used for the transmission measurements performed at GELINA.

Figure 2 Cerium metal sample of 9.93 mm thickness before being encapsulated.

3 Data reduction

The experimental transmission T_{exp} as a function of the time-of-flight was obtained from the ratio of the counts of a sample-in measurement C_{in} and a sample-out measurement C_{out}, after subtraction of the background contributions B_{in} and B_{out}, respectively [1]:

$$T_{\exp}(t_m) = N \frac{C_{in}(t_m) - K\ B_{in}(t_m)}{C_{out}(t_m) - K\ B_{out}(t_m)} \quad , \quad (1)$$

where t_m denotes the measured time-of-flight. The TOF-spectra (C_{in}, B_{in}, C_{out}, B_{out}) in Eq. 1 were corrected for losses due to the dead time in the detector and the electronics chain. All spectra were normalized to the same TOF-bin width structure and to the neutron beam intensity. The latter was derived from the response of the BF_3 beam monitors. To avoid systematic uncertainties due to slow variations of both the beam intensity and the detector efficiency as a function of time, data were taken by alternating sample-in and sample-out measurements in cycles of about 600 seconds each. Such a procedure reduces the uncertainty on the normalization to the beam intensity to less than 0.25 % [1]. This uncertainty was evaluated from the ratios of the counts in the ^{6}Li transmission detector and in the flux monitors. To account for this uncertainty the factor N = 1.0000 $\pm$ 0.0025 was introduced in Eq.1.

The time-of-flight t_m of a neutron creating a signal in the neutron detector was determined by the time difference between the start signal (T_0) and the stop signal (T_s):

$$t_m = (T_s - T_0) + t_0 \quad , \qquad (3)$$

with t_0 a time offset which was determined by a measurement of the γ-ray flash. The flight path distance L = 47.669 (0.004) m, i.e. the distance between the centre of the moderator and the front face of the detector, was derived previously from the result of transmission measurements on ^{238}U using the 6.673 $\pm$ 0.001 eV resonance reported as Derrien *et al.* [9] as reference.

The background as a function of TOF was determined by an analytical expression applying the black resonance technique. The factor K = 1.00 $\pm$ 0.03 in Eq. 1 introduces a correlated uncertainty component accounting for systematic effects due to the background model. Its uncertainty was derived from a statistical analysis on the differences between the estimated background and the observed background in the black resonance dips. The background was parameterized by an analytical expression consisting of a constant and three exponentials [1]:

$$B(t_m) = b_0 + b_1 e^{-\lambda_1 t_m} + b_2 e^{-\lambda_2 t_m} + b_3 e^{-\lambda_3 (t_m + t_0)} \quad . \qquad (2)$$

The time independent contribution b_0 can be estimated from measurements when the accelerator is not in operation. The first exponential accounts for the contribution due to the detection of 2.2 MeV γ-rays resulting from neutron capture in hydrogen that is present in the moderator. The second exponential originates predominantly from neutrons scattered inside the detector station. The last time-dependent component is due to the detection of slow neutrons from previous accelerator cycles. This overlap component was obtained by extrapolating the TOF spectra at the end of the cycle and it was approximated by an exponential, with t0 related to the operating frequency of the acceralator (i.e t_0 = 1.25 ms for 800 Hz). The parameters in the analytical expression were determined by a least squares fit to saturated resonance dips observed in the TOF-spectra resulting from measurements with black resonance filters. The λ_1 and λ_2 parameters were derived from dedicated measurements with Cu, Na, Co, W, Au and Ag black resonance filters in the beam. During the regular sample-in and sample-out runs Co and W fixed black resonance filters were kept in the beam to continuously monitor

the background at 132 eV and 20 eV, respectively, and to account for the dependence of the background level on the presence of the sample [1]. Shorter cycles including an additional Na filter (2.85 keV) were used to study the time dependence of the background (b_1/b_2 ratio). Examples of dead time corrected sample-in TOF-spectra together with the background contributions are shown in Figs. 2 and 3 for the 9.93 mm and the 1.85 mm cerium samples, respectively. In these figures the contributions of the different background components are also given. The parameters of the analytical expression in Eq. 2 for the sample-in and sample-out backgrounds are given for both samples in Table 2 and Table 3.

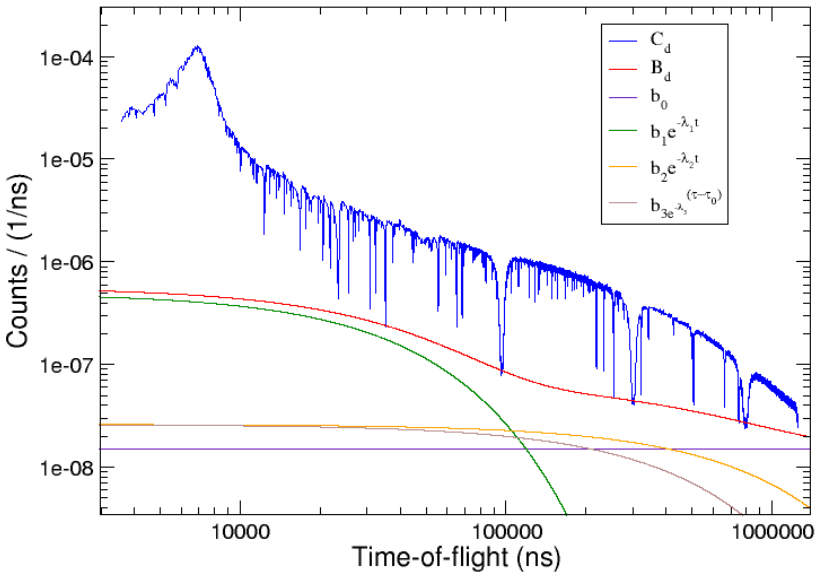

Figure 3 Time-of-flight spectrum obtained with the 9.93 mm thick sample in the beam is compared with the total background and its components.

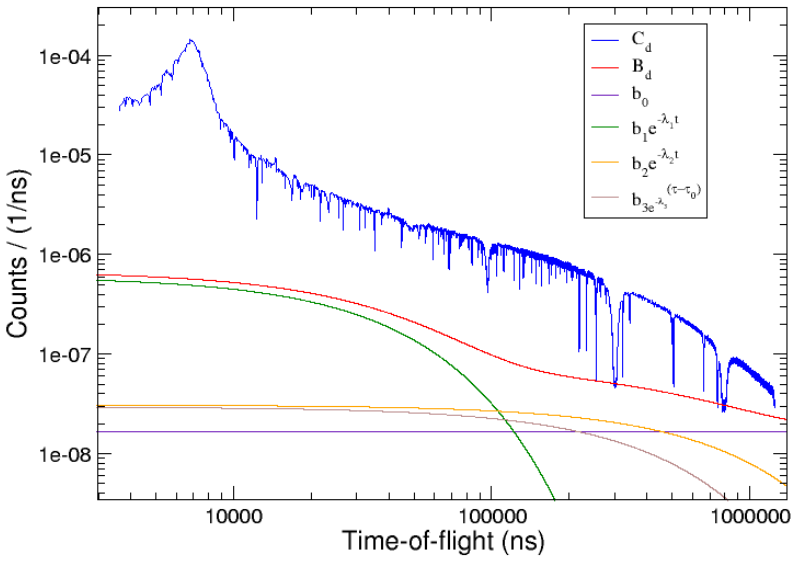

Figure 4 Time-of-flight spectrum with the 1.85 mm thick sample in the beam is compared with the total background and its components.

ID	$b_0/10^{-8}$ ns	$b_1/10^{-7}$ ns	$\lambda_1/10^{-5}$ ns	$b_2/10^{-7}$ ns	$\lambda_2/10^{-6}$ ns	$b_3/10^{-7}$ ns	$\lambda_3/10^{-6}$ ns
C_{in}	1.50	5.06	2.94	0.258	1.35	6.78	2.60
C_{out}	1.77	5.08	2.94	0.292	1.35	7.59	2.60

Table 2 Parameters for the analytical expressions of the background correction for the sample-in and sample-out measurements for the Cerium sample of 9.93 mm.

ID	$b_0/10^{-8}$ ns	$b_1/10^{-7}$ ns	$\lambda_1/10^{-5}$ ns	$b_2/10^{-7}$ ns	$\lambda_2/10^{-6}$ ns	$b_3/10^{-7}$ ns	$\lambda_3/10^{-6}$ ns
C_{in}	1.66	5.58	2.94	0.304	1.35	7.66	2.60
C_{out}	1.68	5.73	2.94	0.315	1.35	7.86	2.60

Table 3 Parameters for the analytical expressions of the background correction for the sample-in and sample-out measurements for the Cerium sample of 1.85 mm.

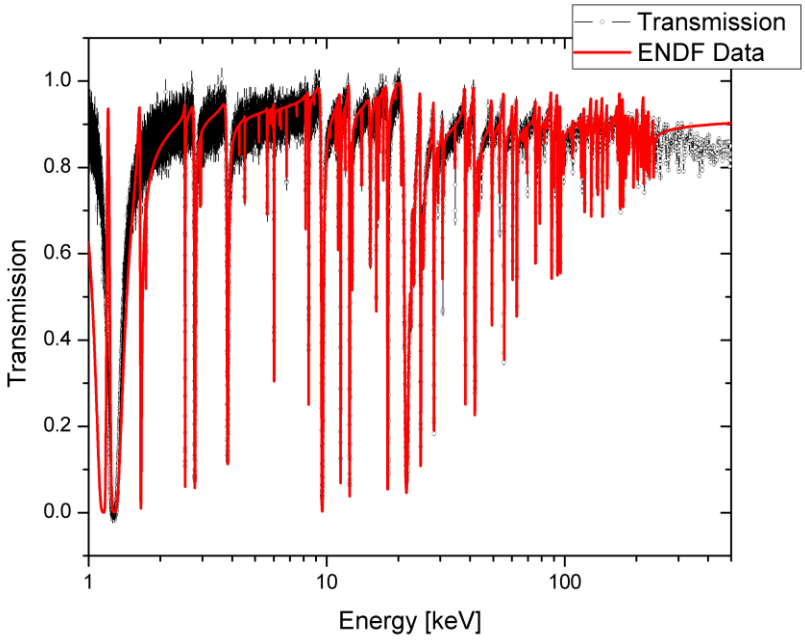

Figure 5 Experimental transmission obtained for the 9.93 mm thick sample is compared to the theoretical transmission obtained when using the ENDF/B-VII.1 total cross section.

4 Results

The experimental transmission is obtained from the ratio of the background subtracted sample-in and sample-out spectra. To derive the experimental transmission and propagate both the correlated and uncorrelated uncertainties the AGS (Analysis of Geel Spectra) code, developed at JRC-Geel, was used [10]. The code is based on a compact formalism to propagate all uncertainties starting from uncorrelated uncertainties due to counting statistics. It stores the full covariance information after each operation in a concise, vectorised way. The AGS formalism results in a substantial reduction of data storage volume and provides a convenient structure to verify the various sources of uncertainties through each step of the data reduction process. The concept is recommended by the Nuclear Data Section of the IAEA [11] to prepare the experimental observables, including their full covariance information, for storage into the EXFOR data library [12].

The experimental transmission resulting from the measurement on the 9.93 mm thick cerium sample is shown in Fig. 4. The format in which the numerical data will be stored in the EXFOR data library is illustrated in Tables B.1 and B.2 in the Appendix. The reported data cover the neutron energy region from 300 eV (above the black resonance region of Co) to 200 keV. The data include the full covariance information based on the AGS concept. The total uncertainty and the uncertainty due to uncorrelated components are reported, together with the contributions due to the normalization (N) and the background model (K). Applying the AGS concept, the covariance matrix V_T of the experimental transmission can be calculated by:

$$V_T = U_u + S(\vec{\eta})S^T(\vec{\eta}), \tag{4.1}$$

where U_u is a diagonal matrix containing the contribution of all uncorrelated uncertainty components. The matrix S is contains the contribution of the components $\vec{\eta} = \{K, N\}$ creating correlated contribution. The uncertainty due to the dead time correction can be neglected.

The experimental details, which are required to perform a resonance analysis on the data, are summarized in Appendix A. The information given is based on the recommendation resulting from a consultant's meeting organized by the NDS-IAEA [5].

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Appendix

A. SUMMARY OF EXPERIMENTAL DETAILS

1.	**Main Reference**		[1,2]
2.	**Facility**	GELINA	[3]
3.	**Neutron production**		
	Neutron production beam	Electron	
	Nominal average beam energy	100 MeV	
	Nominal average peak current	55 μA	
	Repetition rate (pulses per second)	800 Hz	
	Pulse width	1 ns	
	Primary neutron production target	Mercury cooled depleted uranium	
	Target nominal neutron production intensity	3.4 x10^{13} s^{-1}	
4.	**Moderator**		
	Primary neutron source position in moderator	Above and below uranium target	
	Moderator material	2 water filled Be-containers around U-target	
	Moderator dimensions (internal)	2 x (14.6 cm x 21 cm x 3.9 cm)	
	Density (moderator material)	1 g/cm^3	
	Temperature (K)	Room temperature	
	Moderator-room decoupler (Cd, B, …)	None	
5.	**Other experimental details**		
	Measurement type	Transmission	
	Method (total energy, total absorption, …)	Good transmission geometry	[4]
	Flight Path length (m) (centre moderator – detector front face)	L = 47.669(4) m	
	Flight path direction	9° with respect to normal of the moderator face viewing the flight path	
	Neutron beam dimensions at sample position	45 mm in diameter	
	Neutron beam profile	-	
	Overlap suppression	^{10}B overlap filter (8x10^{-3} at/b)	
	Other fixed beam filters	Co, W, Pb (16 mm)	
6.	**Detector**		
	Type	Scintillator (NE912)	
	Material	Li-glass	
	Surface Dimensions	151.6 mm diameter	
	Thickness (cm)	6.35 mm	
	Detector(s) position relative to neutron beam	In the beam	
7.	**Sample**		
	Type (metal, powder, liquid, crystal)	Metal	
	Chemical composition	^{nat}Ce (88.450% ^{140}Ce, 11.114 % ^{142}Ce; 0.251 % ^{138}Ce; 0.185 ^{136}Ce)	
	Sample composition (at/b)	^{nat}Ce (5.534 ± 0.025) 10^{-3} at/b and ^{nat}Ce (28.713 ± 0.025) 10^{-3} at/b	
	Temperature	22°C	
	Sample mass (g)	34.975 g and 187.954 g	
	Geometrical shape (cylinder, sphere, …)	Cylinder	

	Surface dimension	59.85 mm diameter	
	Nominal thickness (mm)	1.85 mm and 9.93 mm	
	Containment description	Al container	
	Additional comment	Transmission measured with respect to a the sample container (dummy sample)	
8.	**Data Reduction Procedure**		[4,5]
	Dead time correction	Done (correction factor < 1.2)	
	Back ground subtraction	Black resonance technique	
	Flux determination (reference reaction, …)	-	
	Normalization	1.000 ± 0.0025	
	Detector efficiency	-	
	Self-shielding	-	
	Time-of-flight binning	Zone length bin width 16384 8 ns 18432 4 ns 8192 8 ns 22528 64 ns	
9.	**Response function**		
	Initial pulse	Normal distribution, FWHM = 2 ns	
	Target / moderator assembly	Numerical distribution from MC simulations	[6,7]
	Detector	Analytical function defined in REFIT manual	[8]

B. Data format

Column	Content	Unit	Comment
1	Energy	eV	Relativistic relation using a fixed flight path length (L = 47.669 m)
2	t_{low}	ns	
3	t_{high}	ns	
4	T_{exp}		Transmission
5	Total Uncertainty		
6	Uncorrelated uncertainty		Uncorrelated uncertainty due to counting statistics
7	AGS-vector (K)		Background model (u_K/K = 3 %)
8	AGS-vector (N)		Normalization (u_N/N = 0.25 %)

Comments from the authors:

- The AGS concept was used to derive the experimental transmission

$$T_{\exp}(t_m) = N\frac{C_{in}(t_m) - K\ B_{in}(t_m)}{C_{out}(t_m) - K\ B_{out}(t_m)}$$

  and to propagate the uncertainties, both the uncorrelated uncertainty due to counting statistics and the uncertainty due to the normalization (u_N/N = 0.25 %) and the background contribution (u_K/K = 3.0 %).

- The quoted uncertainties are standard uncertainties at 1 standard deviation

B.1 DATA (1.85 mm thick sample)

E/ eV	t_l / ns	t_h / ns	T_{exp}	u_t	AGS u_u	AGS K	AGS N
200402.8	7696	7704	0.98169	0.00545	0.00487	0.000002	0.00245
199986.9	7704	7712	0.98208	0.00547	0.00489	0.000002	0.00245
…	…	…	…	…	…		…
1360.097	93448	93456	0.75874	0.02360	0.02355	-0.000492	0.00190
1359.864	93456	93464	0.77811	0.02458	0.02450	-0.000468	0.00194
…	…	…	…	…	…		…
300.0223	198972	198972	0.98957	0.05576	0.05571	-0.000038	0.00247
300.0103	198976	198980	1.01219	0.05685	0.05679	0.000096	0.00253

B.2 DATA (9.93 mm thick sample)

E/ eV	t_l / ns	t_h / ns	T_{exp}	u_t	AGS u_u	AGS K	AGS N
200402.8	7696	7704	0.91919	0.00492	0.00435	-0.00001	0.00230
199986.9	7704	7712	0.91326	0.00491	0.00434	-0.00001	0.00228
…	…	…	…	…	…		…
1360.097	93448	93456	0.25982	0.01204	0.01192	-0.00151	0.00065
1359.864	93456	93464	0.27623	0.01252	0.01230	-0.00147	0.00069
…	…	…	…	…	…		…
300.0223	198972	198972	0.91816	0.05157	0.05151	0.00010	0.00230
300.0103	198976	198980	0.89399	0.05032	0.05027	0.00004	0.00223

Appendix references

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Working in close cooperation with policy Directorates-General, the JRC addresses key societal challenges while stimulating innovation through developing new methods, tools and standards, and sharing its know-how with the Member States, the scientific community and international partners.

Serving society
Stimulating innovation
Supporting legislation

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